

FOOD CHAIN CONCENTRATION OF CHLORINATED HYDROCARBON PESTICIDES IN INVERTEBRATE COMMUNITIES: A RE-EVALUATION

D. M. ROSENBERG*

Department of Entomology

University of Alberta

Edmonton, Alberta T6G 2E3

Quaestiones Entomologicae

11: 97 - 110 1975

Concentration of chlorinated hydrocarbon pesticides in invertebrate food chains is not shown by results reported in the literature. It is proposed that habitat and mode of life rather than trophic level are likely the most important ecological characteristics determining uptake and final concentration of chlorinated hydrocarbon pesticides in aquatic invertebrates and that the idea of "food chain concentration" or "trophic level effect" should be replaced by the idea of "bioconcentration".

Il n'est pas encore démontré au travers des nombreuses publications qu'il y a un rapport chez les invertébrés entre niveau trophique et la concentration des pesticides en hydrocarbures chlorinés. Nous proposons que l'habitat et le mode de vie plutôt que le niveau trophique est à la base de l'absorption et de la concentration finale des hydrocarbures chlorinés chez les invertébrés aquatiques et que l'idée de "la concentration par niveau trophique" ou "l'effet du niveau trophique" soit remplacée par l'idée de "bioconcentration".

INTRODUCTION

A study of the fate of dieldrin in ecosystem components of a slough in central Alberta revealed that the range of concentrations of the pesticide was similar in primary and secondary consumer invertebrates over the duration of the study (Rosenberg, 1975; Table 22). The results of similar studies (Meeks, 1968; Vaajakorpi and Salonen, 1973) revealed the same lack of trophic level effect (Rosenberg, 1975; Table 21).

These results were surprising in view of the generalizations that have existed in the pesticide literature over the last decade or longer regarding the inevitability of food chain concentration of pesticides, especially chlorinated hydrocarbons. Thus, I undertook a literature survey to determine whether or not the results referred to above were atypical.

Because of the immensity of the literature dealing with the effects of pesticides on fauna, the survey dealt almost entirely with the chlorinated hydrocarbons, supposedly the chief offenders in food chain concentration, and mainly considered freshwater invertebrate communities. However, I have given some consideration to pesticide uptake by fish and terrestrial and marine invertebrate communities. In general, only those papers which contained a level of identification sufficient to designate the trophic level to which the animal belonged were used. Other papers are only briefly mentioned.

I have also limited the review to an analysis of the results of field studies and those laboratory studies which have allowed feeding to occur. The use of studies which have allowed feeding is obviously necessary for a consideration of trophic level effects. Studies which have not allowed feeding have already shown that aquatic invertebrates readily concentrate pesticides from water (e.g. Johnson et al., 1971; Wilkes and Weiss, 1971; Derr and Zabik, 1972,

* Present Address: Fisheries & Marine Service, Freshwater Institute, 501 University Crescent, Winnipeg, Manitoba R3T 2N6.

1974). Also, the analysis of studies which have allowed normal trophic relations gives this review a degree of comparability with the studies from which the generalizations about food chain concentration of pesticides originated (e.g. see Rudd, 1964). Admittedly, this approach introduces a degree of circumstantiality into the interpretation. However, in the absence of experiments which quantify the relative contributions made by food and other factors (see below) to the pesticide levels in aquatic invertebrates, I believe the approach is useful.

Variability of residue concentrations in field-collected samples analyzed by gas chromatography is well known (e.g. see Moriarty, 1972; Kenaga, 1972; Rosenberg, 1975, Table 17). Therefore, I have considered differences in residue concentrations from primary to secondary consumer trophic levels to be significant only when these differences are at least an order of magnitude (10-fold).

Finally, I have found that studies of food chain concentrations of pesticides have the following shortcomings which make interpretation of the results difficult and which should be identified at the outset of this review:

1. Changing food habits during a particular life stage and over the lifetime of an animal are usually not considered. For example, different life stages of invertebrates are analyzed simultaneously for pesticide residues. Because specific information on the extent of an animal's mode of feeding prior to residue analysis is usually unavailable, we assume that its major mode of feeding is its only one.
2. Most studies do not correlate a predator with its actual prey. Rather, we assume that of the animals designated as primary consumers some will be eaten by those designated as secondary consumers.
3. The grouping of invertebrates into primary and secondary consumer trophic levels is arbitrary (as are other classification systems of trophic relationships; see Cummins, 1973) and oversimplifies these relationships.

LITERATURE SURVEY

Despite the findings of Godsil and Johnson (1967) that a low concentration of endrin in the lake of their study did not result in food chain concentration, the works of Hunt and Bischoff (1960), Pillmore (in Rudd, 1964), Hunt (in Rudd, 1964), Bridges, Kallman, and Andrews (1963), and Hickey, Keith, and Coon (1966), among others, have reported a trophic level effect resulting from pesticide applications to aquatic ecosystems. Rudd (1964) has discussed instances of trophic level effects in terrestrial ecosystems. However, in each of these studies, invertebrates have been used as a single step in the food chain and usually they are of a single species or are zooplankton. Other studies (Terriere et al., 1966; Keith, 1966; and those reviewed and summarized in Table IV of Moore, 1967) which have also used aquatic invertebrates as a single step, have presumably lumped a number of species of aquatic invertebrates of different trophic levels. Of course, aquatic invertebrates have their own trophic interrelationships (e.g. see Jones, 1949).

Hannon et al. (1970) separated the aquatic invertebrates of their study into three unlikely groups: plankton-algae, crayfish, and aquatic insects (composed of midge larvae and Gyrinidae) so no information on trophic distribution of the chlorinated hydrocarbons is available. Woodwell, Wurster, and Isaacson's (1967) study of the DDT residues in an east coast estuary gives an extensive list of residue data for various species of invertebrates but, unfortunately, none can be classed as secondary consumers. The same is true of the review and summary presented in Table 12 of Edwards (1970) except for the 1964 United States Department of the Interior study which gives a concentration factor for a crab that is the lowest for the entire study. None of the aquatic invertebrates listed in Table 4 of Flickinger and King (1972) are predators except

for the Notonectidae which have been combined with Corixidae under the heading "Aquatic Hemiptera". The range of dieldrin concentrations in the crayfish from Brazoria County in Flickinger and King (1972) spans the concentrations given for all the aquatic invertebrates in Table 4 (none detectable to 17.0 ppm). Also, no secondary consumers are present among the terrestrial invertebrates in Table 10 of Edwards (1970) except for Davis and Harrison's (1966) work which will be considered below.

Moubry, Helm, and Myrdal (1968) reported similar DDT, DDT-metabolites, dieldrin, and endrin levels in *Gammarus* sp., *Limnephilus rhombicus* (L.) larvae, and *Sialis* sp. larvae (Table 1). The last, of course, are predators. Dieldrin residues in some invertebrates exposed to a dieldrin industrial effluent entering the Rocky River, South Carolina, are shown in Table 2 (Wallace and Brady, 1971). It can be seen that the predaceous hellgrammite larvae, *Corydalis cornuta*, had lower levels than the filter-feeding blackfly (*Simulium vittatum*) and caddisfly larvae (*Hydropsyche* sp.). Robinson et al. (1967) reported similar concentrations of DDE and dieldrin in primary and secondary consumer marine invertebrates except in macrozooplankton which they classed as a secondary consumer and which had extraordinarily high concentrations (Table 3). Robinson et al.'s results are shown diagrammatically in Fig. 3 of Edwards (1970). The similarity in pesticide levels between trophic levels 2 (= primary consumer: 0.02 ppm) and 3 (= secondary consumer: 0.03 ppm) is striking. Although Foehrenbach (1972, p. 622) claimed higher dieldrin concentrations existed in invertebrates listed in Table III of his paper than those of the shellfish listed in Table II "... probably because the organisms in Table III are higher in the food chain. . ." the range of concentrations is similar (Table II: 0 to 0.132 mg/kg; Table III: 0.004 to 0.236 mg/kg; the 0.236 value was for a non-predaceous grass shrimp). Also, Table III contains invertebrates that are primary consumers. Naqvi and de la Cruz (1973) analyzed mirex residues in a variety of aquatic, terrestrial, and estuarine invertebrates and vertebrates in Mississippi. In order to calculate mean levels of mirex in herbivore, carnivore, and omnivore trophic levels, the authors combined animals from pond, lake, creek, grassland, and estuarine habitats and lumped invertebrate carnivores with vertebrate carnivores. The mean residue levels presented for these three trophic levels (Table 2 of Naqvi and de la Cruz) and the conclusions reached are, therefore, difficult to interpret. The authors implied food chain concentration occurred even though the increase in mean residues for the three trophic levels was not in the expected order (0.23, 0.30, and 0.35 ppm for herbivores, carnivores, and omnivores respectively). Furthermore, these residue levels are virtually identical in view of the precision possible for samples from the field analyzed by gas-liquid chromatography (see Kenaga, 1972; Moriarty, 1972). However, it is possible to examine whether or not food chain concentration has occurred by using the residue data given for species of aquatic invertebrates. Only residue data for the simultaneous presence of the three trophic levels of aquatic invertebrates is used (Table 4). It can be seen that residue levels in carnivores were highest in only one of four locations. The number of replicates in this location was low for all three trophic levels. Fish (*Gambusia affinis* and *Lepomis cyanellus*) from Bluff Lake had a mean mirex residue of 0.19 ppm (range: 0.07-0.38 ppm; 3 replicates). Fish (same two species as above) from a pond in the Louisville and Noxapater areas had a mean residue of 0.39 ppm (range: 0.17-1.00 ppm; 2 replicates) whereas a single mirex residue in an aquatic invertebrate herbivore was 0.05 ppm and a mean mirex residue of 0.16 ppm (range: 0.07-0.26 ppm; 4 replicates) was present in aquatic invertebrate carnivores in the same area. The single relatively low herbivore residue value is difficult to interpret in view of the ranges of residues reported for aquatic invertebrates in Naqvi and de la Cruz. Collins, Davis, and Markin (1973) reported similar mirex residues in crayfish (range: 0.01 to 0.40 ppm) and dragonfly nymphs (<0.01 to 0.70 ppm).

Table 1. Chlorinated hydrocarbon pesticide residues in invertebrates of Moubry et al.'s (1968) study.

Site	Organism	Residues (ppm)					
		DDE	DDD	DDT	DDT & Analogues	Dieldrin	Endrin
Control	<i>Limnephilus rhombicus</i>	0.014	0.009	0.010	0.033	0.002	
North Branch	<i>Sialis</i> sp.	0.005	0.003	0.008	0.016	0.013	0.009
	<i>Gammarus</i> sp.	0.010	0.007	0.012	0.029	0.003	0.025
	<i>L. rhombicus</i>	0.006	0.007	0.011	0.024	0.002	0.003
Confluence	<i>Gammarus</i> sp.	0.009	0.007	0.015	0.031	0.013	0.013

Table 2. Dieldrin residues in invertebrates of Wallace and Brady's (1971) study*.

Organism	Residue (ppm)+ Position in Relation to Effluent	
	Upstream	Downstream
<i>Simulium vittatum</i>	0.24	16.2
<i>Hydropsyche</i> sp.	0.04	19.0
<i>Corydalis cornuta</i>	0.02	1.2

* Data is for the April 18, 1970 collection, the only date with residue levels for more than a single trophic level.

+ Values shown are means of replicates and the two gas chromatographic columns used.

Table 3. Concentrations of organochlorine compounds in marine invertebrate samples taken off the Northumberland Coast, 1965-1966 (adapted from Robinson et al., 1967).

Organism	Consumer Trophic Level	Concentration (ppm)	
		Dieldrin	pp'-DDE
microzooplankton	1	0.020	0.030
sea urchin (<i>Echinus esculentus</i>)	1	0.027	0.050
mussel (<i>Mytilus edulis</i>)	1	0.023	0.024
cockle (<i>Cardium edule</i>)	1	0.018	0.012
limpet (<i>Patella vulgata</i>)	1	0.009	0.003
macrozooplankton (<i>Crustacea</i>)	2	0.16	0.16
lobster (<i>Homarus vulgaris</i>)	2	0.024	0.024
shore crab (<i>Carcinus maenas</i>)	2	0.025	0.037
edible crab (<i>Cancer pagurus</i>)	2	0.015	0.061

Table 4. Mirex residues in aquatic invertebrates of Naqvi and de la Cruz's (1973) study.

Location ⁺	Residue (ppm) [*]		
	Herbivore	Trophic Level Carnivore	Omnivore
Bluff Lake	0.13 (0.10-0.15) 2	0.47 (0.15-0.78) 2	0.06 (0.04-0.09) 3
Starkville Area, pond	0.13 (0-0.36) 3	0.45 (0.09-1.92) 6	0.45 (0-2.09) 10
Starkville Area, lake	0 1	0.24 (0.02-0.67) 3	1.33 1
Louisville and Noxapater Areas, creek	1.01 (0.12-2.01) 3	0.10 (0-0.23) 7	0.41 (0.02-1.76) 6

+ All Mississippi.

* Mean value is followed by range in parentheses, and number of replicates.

Thus far, only the results of field studies have been discussed. Many laboratory studies have been done on the uptake and accumulation of chlorinated hydrocarbon pesticides by invertebrates but most of these have not allowed feeding. Only the model ecosystem studies (Metcalf, Sangha, and Kapoor, 1971) attempt to duplicate a field situation. Of the many model ecosystem studies consulted, only that of Sanborn and Yu (1973) has used more than one trophic level of invertebrates. Concentrations of dieldrin were highest in the snail (*Physa* sp. – 229.87 ppm), followed by alga (*Oedogonium cardiacum* – 14.96 ppm), fish (*Gambusia affinis* – 12.29 ppm), *Daphnia* sp. (5.07 ppm), *Elodea* sp. (2.56 ppm), clam (*Corbicula manilensis* – 2.03 ppm), and crab (*Uca minax* – 0.495 ppm). In fact, results of the model ecosystem studies of Kapoor et al. (1972, 1973), using an alga (*Oedogonium cardiacum*) – snail (*Physa* sp.) – mosquito (*Culex pipiens quinquefasciatis*) – fish (*Gambusia affinis*) food chain do not follow the classical concept of food chain accumulation (Table 5).

Table 5. Concentrations of chlorinated hydrocarbons in food chain elements of Kapoor et al. (1972, 1973) model ecosystem studies.

Reference	Compound	Concentration (ppm) [*]			
		<i>Oedogonium</i>	<i>Physa</i>	<i>Culex</i>	<i>Gambusia</i>
1972	³ H-Ethoxychlor	2.014	86.16	1.138	4.806
	¹⁴ C-Methylchlor	5.525	101.000	1.002	0.684
1973	[³ H]methoxy-methiochlor	0.074	3.61	1.19	0.15
	[³ H]methyl-ethoxychlor	38.84	10.83	0.74	0.24
	[¹⁴ C]chloro-methylchlor	3.38	31.80	22.94	2.88

* Concentrations are total values for each compound.

There is some evidence that findings of an apparent lack of food chain concentration of chlorinated hydrocarbons (and closely related pesticides) can be extended to fish occupying primary and secondary consumer trophic levels. A number of relatively recent papers have reported that fish do not exhibit food chain concentrations of chlorinated hydrocarbon pesticides. Fredeen, Saha, and Royer (1971) found no difference in the concentration of organochlorine residues in fishes at the end of the food chain (walleyes, saugers, and northern pike) and those lower down (white and longnose suckers, northern redhorse, burbot, goldeye, and yellow perch). Levels of DDT and DDD ranged from < 0.01 to 0.05 ppm, of DDE from < 0.01 to 0.06 ppm, and of dieldrin from 0.002 to 0.006 ppm in fish from all trophic levels. Morris and Johnson (1971) found concentrations of 1600 ppb dieldrin in channel catfish. Other rough fish (buffalo, carp, and carp suckers) had concentrations ranging from 15 to 840 ppb dieldrin while the pan and gamefish (largemouth bass, black and white crappie, black bullhead, bluegill, walleye, and northern pike) had the lowest concentrations (11 to 175 ppb). This latter group, of course, contains several "top predators". Hughes and Lee (1973) concluded that either a clear delineation of toxaphene levels was absent between prey and predator fish or prey fish accumulated higher toxaphene concentrations than the predators (e.g. results from Fox Lake, 6 months after stocking: bluegill and sucker: 9.4 to 10.6 $\mu\text{g/g}$; bass, northern pike, and walleye: 2.2 , 2.3 , and 1.2 $\mu\text{g/g}$). The results of the study of Frank et al. (1974) remind us that factors other than feeding habits are involved in the accumulation of chlorinated hydrocarbons in fish. DDT and dieldrin levels in fish taken from a variety of locations in Ontario, Canada tended to depend on fat content and weight (age) of the fish which, in turn, are related to feeding habits. Risebrough and deLappe (1972) reported that polychlorinated biphenyl (PCB) levels on a fresh weight basis in Atlantic herring were an order of magnitude higher than levels in cod although the latter occupies a higher trophic level. PCB concentrations were comparable on a fat basis (Table 6). Risebrough and deLappe (1972, p. 43) concluded that "the amounts and kinds of lipids may affect the retention of PCB's, modifying the trophic accumulation predicted by the classical food chain concentration theory. Consistent with this hypothesis are the higher PCB residues measured in extractable lipids of the North Atlantic plankton than in the lipids of fish from the same area."

Table 6. Levels of PCB in Atlantic herring and cod (from Risebrough and deLappe, 1972).

Locality	Species	PCB Concentration (ppm)	
		Fresh weight	Extracted lipid
Nova Scotia	Atlantic herring	$0.32-0.54$	
	cod	0.02	
Baltic Sea	Atlantic herring	0.27	6.8
	cod	0.03	11.0

The number of adequate field studies on uptake of pesticides in different trophic levels of terrestrial invertebrates is equally as low as for aquatic invertebrates. The residue values presented by El Sayed, Graves, and Bonner (1967) show conflicting patterns probably because of a lack of collection consistency more than anything else. However, the studies of Davis and Harrison (1966) and Davis (1968) have shown that "... worms and slugs usually contain higher amounts and a greater range of organochlorine compounds than beetles" (Davis, 1968; p. 43 to 44). The beetles he referred to were mostly Carabidae as well as some Staphylinidae and Elateridae. Carabidae and Staphylinidae are predatory. Korschgen (in

Dustman and Stickel, 1969) reported similar levels of dieldrin in earthworms, crickets, and carabids in a Missouri field which had had long-term aldrin applications. Gish (1970) analyzed earthworms of the genera *Allolobophora*, *Diplocardia*, *Helodrilus*, and *Lumbricus*; white grubs (Scarabaeidae larvae); slugs belonging to the genera *Deroceras* and *Limax*; and unidentified terrestrial snails. Unfortunately, he did not analyze any predators. Average concentrations of chlorinated hydrocarbons were 0.6 ppm for the Scarabaeidae larvae, 3.5 ppm for snails (shells included), 13.8 ppm in earthworms, and 89.0 ppm in slugs. Highest levels were 180 times the lowest levels, all for non-predatory forms. Gish (1970) credited the differences in chlorinated hydrocarbon levels to dissimilar feeding habits. Table VII of the review paper by Edwards and Thompson (1973) summarized the results of a large number of pesticide analyses of earthworms, slugs, and beetles (mostly Carabidae). The results are summarized in Table 7. It can be seen that the range of concentrations for the beetles never exceeds that of the earthworms and slugs, something expected of a trophic level effect.

Table 7. Range of concentrations of DDT and dieldrin in earthworms, slugs, and beetles in Table VII of Edwards and Thompson (1973).

Invertebrate Group	Range of Concentrations (ppm)	
	DDT	Dieldrin
earthworms	trace - 680.0	0.06 - 4.6
slugs	0 - 53	0.3 - 18.9
beetles	0.06 - 5.2	0.06 - 9.33

Kenaga (1972) has noted that chlorinated hydrocarbon pesticides best illustrate the mechanisms of bioconcentration of heavy metals and other environmental contaminants. Nonetheless, Moriarty (1972) stated that similar conclusions regarding the lack of a trophic level effect have sometimes been reached for mercury and radionuclides. The literature on uptake and accumulation of heavy metals (e.g. mercury and arsenic) is inconsistent on this point and a review is outside the scope of this paper. By way of speculation, however, it would be surprising if uptake and accumulation of trace and heavy metals by invertebrates was mainly due to a trophic level effect.

Thus, it appears that of the studies which have adequately dealt with chlorinated hydrocarbon pesticide residues in different trophic levels of aquatic or terrestrial invertebrates, a trophic level effect or food chain concentration has not been adequately demonstrated. Yet, unqualified generalizations about food chain concentration of pesticides keep appearing in the literature (e.g. Moore, 1967, p. 113; Dimond, 1969, p. 2, 6; Wurster, 1969, p. 125; Wilkes and Weiss, 1971, p. 223; Foehrenbach, 1972, p. 619, 622, 623, 624; Khan et al., 1973, p. 159, 166; Leland, Bruce, and Shimp, 1973, p. 833; Metcalf, 1973, p. 512).

In his review, Kenaga (1973, p. 80) concluded that "Maximum pesticide residues may sometimes be accumulated by algae or by similar 'first link' organisms in the chain-of-life organisms and do not necessarily result in increasing concentrations in each succeeding link of the chain." Moriarty (1972), in his review of the effects of pesticide residues on wildlife has severely criticized the concept of food chain accumulation of pesticides and other toxic chemicals. He concluded (p. 267): "It is unlikely that predators accumulate the insecticide contained in their prey. All the evidence suggests that aquatic predators acquire their insecticide directly from the water, not from their food." He questioned the validity of a trophic level effect in any food chain. The results and discussion here would lend support to his

contention.

Aquatic organisms acquire pesticides from their surroundings and through their food according to Moore (1967), Chadwick and Brocksen (1969), Dustman and Stickel (1969), Macek (1969), Edwards (1970), Cope (1971), Hamelink, Waybrant, and Ball (1971), Kawatski and Schmulbach (1971), Wilkes and Weiss (1971), Moriarty (1972), and Booth, Yu, and Hansen (1973). To speak of a trophic level effect automatically assumes that food is the more important of the two. It seems likely that habitat and mode of life rather than trophic level are the most important ecological characteristics determining uptake and final concentration of chlorinated hydrocarbon pesticides in aquatic invertebrates. For example, invertebrates leading a planktonic existence usually accumulate the highest concentrations because they provide a lipid source in the water column on which the hydrophobic chlorinated hydrocarbons can adsorb. Once the chlorinated hydrocarbon pesticide has left the water and entered into an organic reservoir it would then be available largely to those organisms associated with the particular substrates for which the chemical has the greatest affinities (Macek, 1969). Invertebrates occupying or contacting a substrate that is high in organic matter favoring partitioning of non-polar pesticides will likely carry high concentrations of chlorinated hydrocarbons (Wallace and Brady, 1971; Derr and Zabik, 1972; Frank et al., 1974). Thus the chemical would not be equally available to all trophic levels in an aquatic community (Macek, 1969). The relatively high concentrations of dieldrin in the *Lymnaea stagnalis*, Chironomidae, Glossiphoniidae, and Libellulidae in Rosenberg (1975) can be explained in this way although other reasons must be sought for the relatively high residues detected in invertebrates not associated primarily with the bottom sediments. Nowhere is the influence of habitat on pesticide uptake better illustrated than in fish. For example, Morris and Johnson (1971) found that bottom-dwelling fish (channel catfish) contained much higher concentrations of dieldrin than non-bottom-dwelling fish (largemouth bass and bluegill). It is not hard to imagine why terrestrial invertebrates such as earthworms and slugs which are constantly in contact with the soil — a potentially large pesticide reservoir — can accumulate higher concentrations than an invertebrate predator which may live in leaf litter and/or scurry around above the soil (e.g. Carabidae).

In terms of the influence mode of life has on pesticide uptake, bivalve molluscs such as oysters, mussels, and marsh clams are well known for their abilities to accumulate high concentrations of chlorinated hydrocarbons (e.g. see Butler, 1969; Khan et al., 1972; Bedford and Zabik, 1973; Petrocelli, Hanks, and Anderson, 1973). Bivalves filter large volumes of water and if the water contains chlorinated hydrocarbons and suspended organic particles to which chlorinated hydrocarbons are adsorbed, the bivalves will contact large amounts of pesticide. However, Bedford and Zabik (1973) have noted that, for mussels, other ecological factors such as: (1) previous conditioning and insecticide residue burden of the mussel; (2) food content and temperature of the water; (3) water quality (including the presence of chemical pollutants and suspended sediment load); and (4) type of pesticide are all involved in the final concentration achieved. Some evidence exists that several trace elements can be magnified in passing from the food to the feces of marine primary consumers (Boothe and Knauer, 1972). Contamination of coastal waters by trace and heavy metals apparently may be as widespread as by pesticides and since fecal material is important to the trophic relationships of coastal benthic communities, the concentrating mechanism may have a significant influence on levels of trace and heavy metals in coprophagous and other members of detrital food webs (Boothe and Knauer, 1972). There is every likelihood that similar processes occur among freshwater invertebrates with chlorinated hydrocarbon pesticides. Dindal and Wurzinger (1971) using the terrestrial snail *Cepaea hortensis* (Müller), showed that highest DDT residues occurred in the feces. They pointed out that since snails frequently re-ingest their own feces, the pesticide can be recycled. Davis (1971) has further illustrated how habitat and mode of life affects

uptake and accumulation of chlorinated hydrocarbons in invertebrates in his consideration of DDT and dieldrin dynamics in two species of earthworms (*Lumbricus terrestris* L. and *Allobophora caliginosa* Sav.). The main factors affecting uptake and accumulation of dieldrin and DDT were: (1) different soil types – Organic matter content will influence the amount of pesticide stored; soil moisture and pH will affect the physiological state of the earthworm and hence its ingestion activity; (2) feeding habits of different species – *A. caliginosa* ingests relatively more soil than *L. terrestris* and at a greater depth. *L. terrestris* is thus more likely to accumulate residues remaining on the vegetation and soil surface. Unfortunately, a paucity exists of this kind of precise habitat and mode of life information as related to pesticides in freshwater invertebrates. It would be more profitable to attempt such studies rather than doing interminable monitoring studies which are already over-abundant in the literature.

Hamelink et al. (1971) have proposed that exchange equilibria control the degree of accumulation of chlorinated hydrocarbons by organisms in lentic environments. This is supported by the earlier findings of Reinert (1967) and Chadwick and Brocksen (1969) that the major uptake of DDT by *Daphnia* and dieldrin by *Cottus perplexus* Gilbert and Evermann respectively was from water and not food. (See also Edwards, 1970). Crosby and Tucker (1971) minimized the importance of ingestion as a route by which *Daphnia* are exposed to suspended chemicals. Derr and Zabik (1974) proposed an adsorption-diffusion mechanism was responsible for the uptake and concentration of DDE by *Chironomus tentans* Fabricius. Macek (1969) and Macek and Korn (1970) reported that brook trout accumulated 10 times more DDT from food than from water in their laboratory studies using approximately 3 ppb. They concluded that since a higher concentration of DDT exists in the food web than in water in natural conditions that the food web is the major source of DDT contamination in fish. Macek (1969) suggested that this was true of lower trophic levels (presumably invertebrates) as well. Macek and Korn's (1970) conclusion that more DDT is available from the food web than from water is not only unsubstantiated by them but also ignores other non-food web reservoirs of DDT in aquatic ecosystems (e.g. suspended organic matter and bottom sediments). Moreover, Murphy (1971) has shown that the results of Macek and Korn (1970) were an artifact of the size of the test fish used. Complexity of the relationship between feeding habits, fat content, and age of fish is illustrated by the study of Frank et al. (1974).

Exchange equilibria would depend on a number of factors according to Hamelink et al. (1971): (1) original concentration of the pesticide in the water (see also Macek et al., 1972; Derr and Zabik, 1972); (2) the water or fat solubility of the pesticide – an increase in water solubility or decrease in fat solubility should reduce the degree of accumulation; (3) the fat content of the animal (see also Morris and Johnson, 1971; Hughes and Lee, 1973; Frank et al., 1974); (4) the species sampled; (5) the time available for exchange (see also Chadwick and Brocksen, 1969; Cope, 1971; Johnson et al., 1971; Wilkes and Weiss, 1971; Morris and Johnson, 1971; Derr and Zabik, 1972); and (6) habitat – persistent pesticides would be tied up in various reservoirs (e.g. algae, bottom sediments of high organic matter content) in a eutrophic lake more so than in an oligotrophic one and thus be less available for accumulation. Size (body weight and surface to volume ratio) of the organism would also be involved (Morris and Johnson, 1971; Murphy, 1971; Kenaga, 1973; Frank et al., 1974). Moriarty (1972) has criticized Hamelink et al.'s (1971) model as being too simplistic and has quite rightly pointed out that it is not the final explanation to the observed phenomena. Nevertheless, in my view, it has been an important contribution.

Other factors, not all of which can be named here, influence the accumulation of pesticide residues by aquatic invertebrates¹. Kenaga (1972, p. 195) has emphasized that the phenomenon

1. For example, see Derr and Zabik's (1974) remarks about the possible role of the epicuticular lipid layer in invertebrates. Also see Wallace and Brady (1971) for possibility of seasonal effects.

is hard to define because of the "many variable inputs, interpretations, and lack of definition in the literature." In trying to explain the lack of consistent food chain buildup in their study, Robinson et al. (1967) wrote of the possibility of a differential ability of vertebrates and invertebrates to metabolize and excrete the pesticide. Pharmacokinetics greatly influence the pesticide levels in organisms (Moore, 1967; Stickel, 1968; Chadwick and Brocksen, 1969; Dustman and Stickel, 1969; Moriarty, 1969, 1972; Edwards, 1970; Cope, 1971; Hamelink et al., 1971; Kawatski and Schmulbach, 1971; and Wilkes and Weiss, 1971). Some pesticide accumulation undoubtedly results from ingestion (Kenaga, 1973). Physical and chemical properties of the pesticides, in addition to solubility and partitioning coefficients already discussed, are involved (see Kenaga, 1972, 1973). Finally, extrinsic factors must affect pesticide levels in aquatic invertebrates: (1) varying concentrations (e.g. see Chadwick and Brocksen, 1969; Hamelink et al., 1971; Wilkes and Weiss, 1971); and (2) whether exposure is acute or chronic (e.g. see Moore, 1967; Stickel, 1968; Chadwick and Brocksen, 1969; Moriarty, 1969; Edwards, 1970; Johnson et al., 1971).

It is important to realize that a literature survey which shows an apparent absence of a trophic level effect in invertebrate communities is only indirect evidence that food chain uptake is not as important as other ecological factors in determining the concentrations of pesticides in invertebrates. Determination of the relative, quantitative contributions made by the factors discussed above to the pesticide levels achieved in invertebrates depends on carefully controlled experimentation and is a critical research need in the study of pesticide-fauna interactions. Until such research is done, and perhaps even after, we should not be using the terms "food chain concentration" or "trophic level effect". Rather, we should talk of "bioconcentration" which Kenaga (1973, p. 75) has defined as "the amount of a pesticide residue accumulated by an organism by adsorption, and by absorption via oral or other route of entry, which results in an increased concentration of the pesticide by the organisms or specific tissues."

SUMMARY AND CONCLUSIONS

1. Concentrations of chlorinated hydrocarbon pesticides in aquatic invertebrates as reported in the literature do not reveal a trophic level effect.
2. Uptake and accumulation of chlorinated hydrocarbon pesticides in aquatic invertebrates is more likely a function of habitat, mode of life, and exchange equilibria than food but is also affected by size of the organism, pharmacokinetics, physical and chemical properties of the pesticides, and various extrinsic factors.
3. Until adequate research is done, the relative contributions of the factors listed above to pesticide levels in invertebrates will remain unknown.
4. The idea of "food chain concentration" or "trophic level effect" is inaccurate and should be replaced by the more accurate idea denoted by the term "bioconcentration".

ACKNOWLEDGEMENTS

I would like to thank W. G. Evans for his advice, continual encouragement, and the many helpful discussions we have had. Thanks are also due to the other members of my advisory committee for their help in producing the thesis which formed the basis of this paper: G. E. Ball, Department of Entomology, University of Alberta; H. F. Clifford and J. R. Nursall, both of the Department of Zoology, University of Alberta; and W. F. Allen, Department of Chemistry, University of Alberta. F. A. J. Armstrong, M. Healey, and R. D. Hamilton of the Freshwater Institute, Winnipeg, G. R. B. Webster, Pesticide Research Unit, University of Manitoba, and D. Pimentel, Cornell University, critically read the manuscript. I have used many of their suggestions.

G. Porth and C. Plumridge, Freshwater Institute, Winnipeg, typed drafts of the manuscript. A. P. Wiens and C. Madder helped proofread the drafts. E. Marshall and his staff (Freshwater Institute Library) provided help obtaining some of the literature.

This research was supported by National Research Council of Canada and Canadian Wildlife Service grants to Dr. W. G. Evans. I am pleased to acknowledge my appreciation to the National Research Council of Canada for scholarships awarded.

REFERENCES

- Bedford, J. W. and M. J. Zabik. 1973. Bioactive compounds in the aquatic environment: uptake and loss of DDT and dieldrin by freshwater mussels. *Archives of Environmental Contamination and Toxicology* 1: 97-111.
- Booth, G. M., C.-C. Yu, and D. J. Hansen. 1973. Fate, metabolism, and toxicity of 3-isopropyl-1H-2, 1,3-benzothiadiazin-4 (3H)-1-2, 2-dioxide in a model ecosystem. *Journal of Environmental Quality* 2: 408-411.
- Boothe, P. N. and G. A. Knauer. 1972. The possible importance of fecal material in the biological amplification of trace and heavy metals. *Limnology and Oceanography* 17: 270-274.
- Bridges, W. R., B. J. Kallman, and A. K. Andrews. 1963. Persistence of DDT and its metabolites in a farm pond. *Transactions of the American Fisheries Society* 92: 421-427.
- Butler, P. A. 1969. The sub-lethal effects of pesticide pollution, p. 87-89. *In* J. W. Gillett (Editor). *The biological impact of pesticides in the environment*. Environmental Health Sciences Series No. 1.
- Chadwick, G. G. and R. W. Brocksen. 1969. Accumulation of dieldrin by fish and selected fish-food organisms. *Journal of Wildlife Management* 33: 693-700.
- Collins, H. L., J. R. Davis, and G. P. Markin. 1973. Residues of Mirex in channel catfish and other aquatic organisms. *Bulletin of Environmental Contamination and Toxicology* 10: 73-77.
- Cope, O. B. 1971. Interactions between pesticides and wildlife. *Annual Review of Entomology* 16: 325-364.
- Crosby, D. G. and R. K. Tucker. 1971. Accumulation of DDT by *Daphnia magna*. *Environmental Science and Technology* 5: 714-716.
- Cummins, K. W. 1973. Trophic relations in aquatic insects. *Annual Review of Entomology* 18: 183-206.
- Davis, B. N. K. 1968. The soil macrofauna and organochlorine insecticide residues at twelve agricultural sites near Huntingdon. *Annals of Applied Biology* 61: 29-45.
- Davis, B. N. K. 1971. Laboratory studies on the uptake of dieldrin and DDT by earthworms. *Soil Biology and Biochemistry* 3: 221-233.
- Davis, B. N. K. and R. B. Harrison. 1966. Organochlorine insecticide residues in soil invertebrates. *Nature, London* 211: 1424-1425.
- Derr, S. K. and M. J. Zabik. 1972. Biologically active compounds in the environment: The uptake and distribution of [1, 1-dichloro-2,2-bis (p-chlorophenyl) ethylene], DDE by *Chironomus tentans* Fabricius (Diptera: Chironomidae). *Transactions of the American Fisheries Society* 101: 323-329.
- Derr, S. K. and M. J. Zabik. 1974. Bioactive compounds in the aquatic environment: Studies on the mode of uptake of DDE by the aquatic midge, *Chironomus tentans* (Diptera: Chironomidae). *Archives of Environmental Contamination and Toxicology* 2: 152-164.
- Dimond, J. B. 1969. DDT in Maine forests. A report to the 104th Maine Legislature. Maine Agricultural Experiment Station Miscellaneous Report Number 125. 13 p.
- Dindal, D. L. and K.-H. Wurzinger. 1971. Accumulation and excretion of DDT by the terrestrial snail *Cepaea hortensis*. *Bulletin of Environmental Contamination and Toxicology* 6: 362-371.

- Dustman, E. H. and L. F. Stickel. 1969. The occurrence and significance of pesticide residues in wild animals. *Annals of the New York Academy of Sciences* 160: 162-172.
- Edwards, C. A. 1970. Persistent pesticides in the environment. Chemical Rubber Company (Cleveland), *Critical Reviews in Environmental Control* 1: 7-68.
- Edwards, C. A. and A. R. Thompson. 1973. Pesticides and the soil fauna. *Residue Reviews* 45: 1-79.
- El Sayed, E. I., J. B. Graves, and F. L. Bonner. 1967. Chlorinated hydrocarbon insecticide residues in selected insects and birds found in association with cotton fields. *Journal of Agricultural and Food Chemistry* 15: 1014-1017.
- Flickinger, E. L. and K. A. King. 1972. Some effects of aldrin-treated rice on Gulf Coast wildlife. *Journal of Wildlife Management* 36: 706-726.
- Foehrenbach, J. 1972. Chlorinated pesticides in estuarine organisms. *Journal of the Water Pollution Control Federation* 44: 619-624.
- Frank, R., A. E. Armstrong, R. G. Boelens, H. E. Braun, and C. W. Douglas. 1974. Organochlorine insecticide residues in sediment and fish tissues, Ontario, Canada. *Pesticides Monitoring Journal* 7: 165-180.
- Fredeen, F. J. H., J. G. Saha, and L. M. Royer. 1971. Residues of DDT, DDE, and DDD in fish in the Saskatchewan River after using DDT as a blackfly larvicide for twenty years. *Journal of the Fisheries Research Board of Canada* 28: 105-109.
- Gish, C. D. 1970. Organochlorine insecticide residues in soils and soil invertebrates from agricultural lands. *Pesticides Monitoring Journal* 3: 241-252.
- Godsil, P. J. and W. C. Johnson. 1967. Pesticide monitoring of the aquatic biota at the Tule Lake National Wildlife Refuge. *Pesticides Monitoring Journal* 1: 21-26.
- Hamelink, J. L., R. C. Waybrant, and R. C. Ball. 1971. A proposal: exchange equilibria control the degree chlorinated hydrocarbons are biologically magnified in lentic environments. *Transactions of the American Fisheries Society* 100: 207-214.
- Hannon, M. R., Y. A. Greichus, R. L. Applegate, and A. C. Fox. 1970. Ecological distribution of pesticides in Lake Poinsett, South Dakota. *Transactions of the American Fisheries Society* 99: 496-500.
- Hickey, J. J., J. A. Keith, and F. B. Coon. 1966. An exploration of pesticides in a Lake Michigan ecosystem. *Journal of Applied Ecology* 3 (Supplement): 141-154.
- Hughes, R. A. and G. F. Lee. 1973. Toxaphene accumulation in fish in lakes treated for rough fish control. *Environmental Science and Technology* 7: 934-939.
- Hunt, E. G. and A. I. Bischoff. 1960. Inimical effects on wildlife of periodic DDD applications to Clear Lake. *California Fish and Game* 46: 91-106.
- Johnson, B. T., C. R. Saunders, H. O. Sanders, and R. S. Campbell. 1971. Biological magnification and degradation of DDT and aldrin by freshwater invertebrates. *Journal of the Fisheries Research Board of Canada* 28: 705-709.
- Jones, J. R. E. 1949. A further ecological study of a calcareous stream in the "Black Mountain" district of south Wales. *Journal of Animal Ecology* 18: 142-159.
- Kapoor, I. P., R. L. Metcalf, A. S. Hirwe, P.-Y. Lu, J. R. Coats, and R. F. Nystrom. 1972. Comparative metabolism of DDT, methylchlor, and ethoxychlor in mouse, insects, and in a model ecosystem. *Journal of Agricultural and Food Chemistry* 20: 1-6.
- Kapoor, I. P., R. L. Metcalf, A. S. Hirwe, J. R. Coats, and M. S. Khalsa. 1973. Structure activity correlations of biodegradability of DDT analogs. *Journal of Agricultural and Food Chemistry* 21: 310-315.
- Kawatski, J. A. and J. C. Schmulbach. 1971. Accumulation of insecticide in freshwater ostracods exposed continuously to sublethal concentrations of aldrin or dieldrin. *Transactions of the American Fisheries Society* 100: 565-567.

- Keith, J. O. 1966. Insecticide contaminations in wetland habitats and their effects on fish-eating birds. *Journal of Applied Ecology* 3 (Supplement): 71-85.
- Kenaga, E. E. 1972. Factors related to the bioconcentration of pesticides, p. 193-228. *In* F. Matsumura, G. M. Bousch, and T. Misato. (Editors). *Environmental toxicology of pesticides*. Academic Press, New York.
- Kenaga, E. E. 1973. Guidelines for environmental studies of pesticides: Determination of bioconcentration potential. *Residue Reviews* 44: 73-113.
- Khan, M. A. Q., A. Kamal, R. J. Wolin, and J. Runnels. 1972. *In vivo* and *in vitro* epoxidation of aldrin by aquatic food chain organisms. *Bulletin of Environmental Contamination and Toxicology* 8: 219-228.
- Khan, M. A. Q., R. H. Stanton, D. J. Sutherland, J. D. Rosen, and N. Maitra. 1973. Toxicity-metabolism relationship of the photoisomers of certain chlorinated cyclodiene insecticide chemicals. *Archives of Environmental Contamination and Toxicology* 1: 159-169.
- Leland, H. V., W. N. Bruce, and N. F. Shimp. 1973. Chlorinated hydrocarbon insecticides in sediments of southern Lake Michigan. *Environmental Science and Technology* 7: 833-866.
- Macek, K. J. 1969. Biological magnification of pesticide residues in food chains, p. 17-21. *In* J. W. Gillett (Editor). *The biological impact of pesticides in the environment*. Environmental Health Sciences Series Number 1.
- Macek, K. J. and S. Korn. 1970. Significance of the food chain in DDT accumulation by fish. *Journal of the Fisheries Research Board of Canada* 27: 1496-1498.
- Macek, K. J., D. F. Walsh, J. W. Hogan, and D. D. Holz. 1972. Toxicity of the insecticide Dursban to fish and aquatic invertebrates in ponds. *Transactions of the American Fisheries Society* 101: 420-427.
- Meeks, R. L. 1968. The accumulation of ^{36}Cl ring-labelled DDT in a freshwater marsh. *Journal of Wildlife Management* 32: 379-396.
- Metcalfe, R. L. 1973. A century of DDT. *Journal of Agricultural and Food Chemistry* 21: 511-519.
- Metcalfe, R. L., G. K. Sangha, and I. P. Kapoor. 1971. Model ecosystem for the evaluation of pesticide biodegradability and ecological magnification. *Environmental Science and Technology* 5: 709-714.
- Moore, N. W. 1967. A synopsis of the pesticide problem, p. 75-129. *In* J. B. Cragg (Editor). Volume 4. *Advances in ecological research*. Academic Press, London.
- Moriarty, F. 1969. The sublethal effects of synthetic insecticides on insects. *Biological Reviews* 44: 321-357.
- Moriarty, F. 1972. The effects of pesticides on wildlife: exposure and residues. *Science of the total Environment* 1: 267-288.
- Morris, R. L. and L. G. Johnson. 1971. Dieldrin levels in fish from Iowa streams. *Pesticides Monitoring Journal* 5: 12-16.
- Moubry, R. J., J. M. Helm, and G. R. Myrdal. 1968. Chlorinated pesticide residues in an aquatic environment located adjacent to a commercial orchard. *Pesticides Monitoring Journal* 1: 27-29.
- Murphy, P. G. 1971. The effect of size on the uptake of DDT from water by fish. *Bulletin of Environmental Contamination and Toxicology* 6: 20-23.
- Naqvi, S. M. and A. A. de la Cruz. 1973. Mirex incorporation in the environment: Residues in nontarget organisms - 1972. *Pesticides Monitoring Journal* 7: 104-111.
- Petrocelli, S. R., A. R. Hanks, and H. W. Anderson. 1973. Uptake and accumulation of an organochlorine insecticide (dieldrin) by an estuarine mollusc, *Rangia cuneata*. *Bulletin of Environmental Contamination and Toxicology* 10: 315-320.
- Reinert, R. E. 1967. The accumulation of dieldrin in an alga (*Scenedesmus obliquus*), *Daphnia*

- (*Daphnia magna*), guppy (*Lebistes reticulatus*) food chain. Dissertation Abstracts 28: 2210-B.
- Risebrough, R. W. and B. deLappe. 1972. Accumulation of polychlorinated biphenyls in ecosystems. *Environmental Health Perspectives* 1: 39-45.
- Robinson, J., A. Richardson, A. N. Crabtree, J. C. Coulson, and G. R. Potts. 1967. Organochlorine residues in marine organisms. *Nature, London* 214: 1307-1311.
- Rosenberg, D. M. 1975. Fate of dieldrin in sediment, water, vegetation, and invertebrates of a slough in central Alberta, Canada. *Quaestiones Entomologicae* 11: 69-96.
- Rudd, R. L. 1964. *Pesticides and the living landscape*. University of Wisconsin Press, Madison. 320 p.
- Sanborn, J. R. and C.-C. Yu. 1973. The fate of dieldrin in a model ecosystem. *Bulletin of Environmental Contamination and Toxicology* 10: 340-346.
- Stickel, L. F. 1968. Organochlorine residues in the environment. United States Fish and Wildlife Service, Special Scientific Report — Wildlife Number 119. 32 p.
- Terriere, L. C., U. Kiigemagi, A. R. Gerlach, and R. L. Borovicka. 1966. The persistence of toxaphene in lake waters and its uptake by aquatic plants and animals. *Journal of Agricultural and Food Chemistry* 14: 66-69.
- Vaajakorpi, H. A. and L. Salonen. 1973. Bioaccumulation and transfer of ^{14}C -DDT in a small pond ecosystem. *Annales Zoologici Fennici* 10: 539-544.
- Wallace, J. B. and U. E. Brady. 1971. Residue levels of dieldrin in aquatic invertebrates and effect of prolonged exposure on populations. *Pesticides Monitoring Journal* 5: 295-300.
- Wilkes, F. G. and C. M. Weiss. 1971. The accumulation of DDT by the dragonfly nymph *Tetragoneuria*. *Transactions of the American Fisheries Society* 100: 222-236.
- Woodwell, G. M., C. F. Wurster, Jr., and P. A. Isaacson. 1967. DDT residues in an east coast estuary: a case for biological concentration of a persistent pesticide. *Science* 156: 821-824.
- Wurster, C. F., Jr. 1969. Chlorinated hydrocarbon insecticides and the world ecosystem. *Biological Conservation* 1: 123-129.